Ageing effects in single particle trajectory averages

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We study time averages of single particle trajectories in scale free anomalous diffusion processes, in which the measurement starts at some time $t_a > 0$ after initiation of the process at the time origin, $t = 0$. Using ageing renewal theory we show that for such non-stationary processes a large class of observables are affected by a unique ageing function, which is independent of boundary conditions or the external forces. We quantify the weakly non-ergodic nature of this process in terms of the distribution of time averages and the ergodicity breaking parameter which both explicitly depend on the ageing time t_a. Consequences for the interpretation of single particle tracking data are discussed.

Ergodicity in the Boltzmann sense, the equivalence of sufficiently long time and ensemble averages of a physical observable, and time translational invariance are hallmarks of many classical systems. In contrast, ergodicity violation and ageing effects are found in the dynamics of various complex systems including glasses [1], blinking quantum dots [2], weakly chaotic maps [3], and single particle tracking experiments in living biological cells [4].

As originally pointed out by Bouchaud [1], when the lifetimes t of states of a physical system are power-law distributed, $\psi(t) \simeq \tau^\alpha/t^{1+\alpha}$ $(0 < \alpha < 1)$, with an infinite mean sojourn time $\langle t \rangle$, both weak ergodicity breaking and ageing in the following sense occur: time averages of physical observables remain random even in the limit of long measurement times and differ from the corresponding ensemble averages [5–14]. Furthermore ensemble averaged correlation functions of observables, taken at two time instants t_2 and t_1, are not solely functions of the time difference $|t_2 - t_1|$. With this breakdown of stationarity, the statistical properties of such systems are no longer time translation invariant. Consider, for example, a particle undergoing a random walk in a random environment, such that the particle hops a finite distance to the left or right with equal probability, but sojourn times between jumps are distributed like $\psi(t)$. Such a model, called the continuous time random walk model (CTRW), was introduced by Scher and Montroll in the context of charge carriers in disordered systems [15]. Its intriguing statistical properties become apparent when studying time averages, like the time averaged mean squared displacement (TAMSD), which is commonly used to analyze the diffusive properties measured in single particle tracking assays. Assume that the process starts at time $t = 0$. From a single trajectory $x(t)$, observed in the time interval $(t_a, t_a + T)$, the TAMSD is typically determined through the definition

$$\overline{\delta^2(\Delta; t_a, T)} = \int_{t_a}^{t_a+T-\Delta} \frac{[x(t + \Delta) - x(t)]^2}{T - \Delta} dt, \quad (1)$$

with the lag time Δ and the measurement time T.

For Brownian motion, in the limit of large T, we obtain the expected behavior, namely, $\overline{\delta^2} \to 2K_1\Delta$. In this case, the process does not age: the result does not depend on the choice of t_a. Moreover Brownian motion is ergodic: the result for the TAMSD is not random, and the same as found for an ensemble of Brownian particles, $\langle x^2(\Delta) \rangle = 2K_1\Delta$ [16]. Thus the diffusion constant K_1 can be determined either from an ensemble of trajectories as originally done by Perrin [17] or, as conceived by Nordlund [18], from a single particle trajectory. For anomalous diffusion, defined in terms of $\langle x^2(\Delta) \rangle = 2K_\alpha\Delta^\alpha/\Gamma(1 + \alpha)$ with the generalized diffusion constant K_α, the equivalence between time and ensemble averages as well as the time translation invariance generally breaks down. One of our central results is that for CTRW processes with long tailed $\psi(t)$ the aged TAMSD (1) for $\Delta \ll T$ follows

$$\left\langle \overline{\delta^2(\Delta; t_a, T)} \right\rangle = \frac{\Lambda_\alpha(t_a/T)}{\Gamma(1 + \alpha)} \frac{g(\Delta)}{T^{1-\alpha}}, \quad (2)$$

where $\Lambda_\alpha(z) = (1 + z)^\alpha - z^\alpha$, and g is a function of Δ only. Another important finding is that $\overline{\delta^2}$ remains a random variable, whose statistical properties explicitly depend on both process age t_a and measurement time T.

Previous work revealed deviations from standard ergodic statistical mechanics by studying time averages in the interval $(t_a = 0, T)$ [6–12]. Namely, in these works the start of the measurement coincides with the start of the process [19]. However, in experiment, the observed particle may be immersed in the medium long before we start our observation. In fact in some cases we may not even know when the process was initiated.

In this Letter, we derive the ageing renewal theory for CTRW processes and study the dependence of time averages of physical observables on the starting time t_a of a measurement after system preparation at $t = 0$. According to Eq. (2), the time intervals $(0, T)$ and $(t_a, t_a + T)$

are not equivalent: In complete contrast to Brownian motion, the statistical properties of CTRW trajectories will depend on the observation window, the process exhibits *ageing*. The remarkable property of Eq. (2) is that corrections due to ageing enter in terms of a unique prefactor depending on t_a and the measurement time T. We call this Λ_α the ageing depression function, which is *independent* of Δ. This function contains all the information on ageing, and is universal since the formula applies for any external force or boundary condition, and, as shown below, also holds for a large class of physical observables. The function $g(\Delta)$ contains the complete Δ-dependence. For instance, we find $g(\Delta) \simeq \Delta$ for free motion [6, 7] and $g(\Delta) \simeq \Delta^{1-\alpha}$ at long times under confinement [9, 10].

Ageing renewal theory. In a CTRW, the position coordinate x of a random walker is an accumulation of random jump lengths, $x(n) = \sum_{i=0}^{n} \delta x_i$. In the simplest, unbiased version of the model, the δx_i are independent, identically distributed (IID) random variables with zero mean, and we assume also finite variance σ^2. Jumps are separated by random IID waiting times, drawn from the common distribution $\psi(t) \simeq \tau^\alpha / t^{1+\alpha}$, with $0 < \alpha < 1$. This implicitly defines a counting process $n(t)$, the number of steps up to time t. The statistics of the overall diffusion process $x(t) = x(n(t))$ are to be derived from the properties of both of its constituents, a method commonly called subordination [20–22]. For a large variety of physical applications of CTRW and non-aged renewal theory see [15, 23, 24]. We first discuss the counting process $n(t)$, emphasizing the role of ageing.

Since waiting times are independent, the counting process $n(t)$ is a renewal process, which we assume to start at $t = 0$. To study the ageing properties of the system, we consider $n_a(t_a, t) = n(t + t_a) - n(t_a)$, the number of renewals in the interval (t_a, t_a+t). The corresponding probability density in double Laplace space, $(t_a, t) \to (s_a, s)$, in the scaling limit of large times becomes [25, 26]

$$p(n_a; s_a, s) = \delta(n_a) \left(\frac{1}{s_a s} - \frac{h(s_a, s)}{s} \right) + \frac{h(s_a, s)}{s^{1-\alpha}} \tau^\alpha e^{-n_a (s\tau)^\alpha}. \quad (3)$$

Here the probability of the waiting time for the first jump to occur after start of the measurement at t_a is [26–28]

$$h(s_a, s) = \frac{s_a^\alpha - s^\alpha}{s_a^\alpha (s_a - s)} \quad \Leftrightarrow \quad h(t_a, t) = \frac{t_a^\alpha}{t^\alpha (t_a + t)}. \quad (4)$$

In the Brownian limit $\alpha \to 1$, the number of jumps n and real time t are equivalent, $p(n_a; t_a, t) = \delta(n_a - t/\tau)$.

This formalism allows for a direct calculation of the average of any function of $n_a(t_a, t)$. For instance, the qth order moment becomes [29]

$$\begin{aligned} \langle n_a^q(s_a, s) \rangle &= \int_0^\infty n_a^q \, p(n_a; s_a, s) \, dn_a \\ &= \Gamma(q+1) \frac{s_a^\alpha - s^\alpha}{s_a^\alpha (s_a - s)} \frac{\tau^\alpha}{s^{1+\alpha q}}. \end{aligned} \quad (5)$$

After double Laplace inversion, we find

$$\begin{aligned} \langle n_a^q(t_a, t) \rangle &= \Gamma(q+1) / [\Gamma(\alpha) \Gamma(1 + \alpha q - \alpha)] \\ &\times \left(\frac{t + t_a}{\tau} \right)^{\alpha q} B \left(\frac{t}{t + t_a}; 1 + \alpha q - \alpha, \alpha \right), \end{aligned} \quad (6)$$

where $B(z; a, b)$ is the incomplete beta function [30]. Thus, the number of steps taken during a time interval of length t is not stationary in distribution: the moments for the period $[0, t]$ are clearly different from those for $[t_a, t_a + t]$. Indeed we see from Eqs. (5) and (6) that the process gets slower and eventually stalls as $t_a \to \infty$. Concurrently the t-dependence changes with t_a: $\langle n_a^q(0, t) \rangle \simeq t^{\alpha q}$ at $t_a = 0$, but $\langle n_a^q(t_a, t) \rangle \simeq t_a^{\alpha-1} t^{1-\alpha+\alpha q}$ for large but finite t_a/t.

For the probability density (3) we obtain

$$\begin{aligned} p(n_a; t_a, t) &\sim [1 - m_\alpha(t/t_a)] \delta(n_a) + m_\alpha(t/t_a) \Gamma(2 - \alpha) \\ &\times \frac{1}{(t/\tau)^\alpha} H_{1,1}^{1,0} \left[\frac{n_a}{(t/\tau)^\alpha} \middle| \begin{array}{c} (2 - 2\alpha, \alpha) \\ (0, 1) \end{array} \right] \end{aligned} \quad (7)$$

for $t_a \gg t$, in terms of an H-function [31, 32] and the probability to make at least one step during $[t_a, t_a + t]$,

$$m_\alpha(t/t_a) = \frac{B\left([1 + t_a/t]^{-1}, 1 - \alpha, \alpha\right)}{\Gamma(1 - \alpha) \Gamma(\alpha)}. \quad (8)$$

Again, we emphasize the explicit dependence on t_a. When compared to the form for $t_a = 0$ [6, 27], the most striking difference in Eq. (7) is the occurrence of the $\delta(n_a)$-term: For a non-aged process ($t_a = 0$), we have $m_\alpha = 1$, while an aged process always has a nonzero probability of not performing any steps at all within the chosen time window, its amplitude approaching 1 algebraically, $m_\alpha \sim (t/t_a)^{1-\alpha} / [\Gamma(\alpha) \Gamma(2 - \alpha)]$, as $t_a \gg t$.

Ageing CTRW. We proceed by adding the position coordinate to our description. For IID jump distances δx_i, the jump process $x(n)$ converges in distribution to free Brownian motion in the scaling limit of many jumps. The anomalous diffusion process $x(t) = x(n(t))$ inherits the ageing properties of the counting process discussed above. To see this, consider the qth order TA moments

$$\left\langle \overline{\delta^q(\Delta; t_a, T)} \right\rangle = \int_{t_a}^{t_a + T - \Delta} \frac{\langle |x(t + \Delta) - x(t)|^q \rangle}{T - \Delta} dt, \quad (9)$$

which are useful to characterize experimental data [33]. For free Brownian motion $x(n)$ we know that

$$\langle |x(n_2) - x(n_1)|^q \rangle = \frac{2\Gamma(q)}{\Gamma(q/2)} \frac{\sigma^q}{2^{q/2}} |n_2 - n_1|^{q/2}, \quad (10)$$

Since $x(n)$ and $n(t)$ are independent processes, we use conditional averaging to evaluate the integrand of Eq. (9). From above result (6), it follows that

$$\begin{aligned} \langle |x(t + \Delta) - x(t)|^q \rangle &= \frac{2\Gamma(q)}{\Gamma(q/2)} \frac{\sigma^q}{2^{q/2}} \langle n_a^{q/2}(t, \Delta) \rangle \\ &= \Gamma(q+1) / [\Gamma(\alpha) \Gamma(1 + \alpha q/2 - \alpha)] \\ &\times [K_\alpha (t + \Delta)^\alpha]^{q/2} B \left(\frac{\Delta}{t + \Delta}, 1 - \alpha + \alpha q/2, \alpha \right) \end{aligned} \quad (11)$$
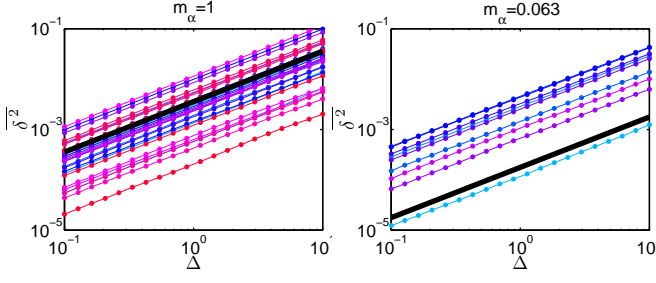
Figure 1: Time averaged mean squared displacement $\overline{\delta^2(\Delta; t_a, T)}$ for individual free CTRW trajectories (full symbols) and the averages according to Eq. (12) (bold black lines). Left: Measurement starts at $t_a = 0$, so that $m_\alpha = 1$. Right: Aged process, $t_a = 10^{11}$ (a.u.), so that a large fraction $1 - m_\alpha \approx 94\%$ of trajectories is suppressed in the log-log plot. The parameters are $\alpha = 1/2$, $\tau = 1$, $\sigma^2 = 1$ and $T = 10^9$.

where we identify $K_\alpha = \sigma^2/(2\tau^\alpha)$ [23]. The qth order TA moment (9) in the limit $\Delta \ll T$ then becomes

$$\left\langle \overline{\delta^q(\Delta; t_a, T)} \right\rangle = \frac{\Lambda_\alpha(t_a/T) [K_\alpha \Delta^\alpha]^{q/2} \Gamma(q+1)}{\Gamma(\alpha+1)\Gamma(2-\alpha+\alpha q/2)} \left(\frac{\Delta}{T}\right)^{1-\alpha},$$
(12)

which is of the general shape (2) with $g(\Delta) \sim \Delta^{1-\alpha+\alpha q/2}$. Interestingly, we see the special role of the TAMSD ($q = 2$), for which the Δ-scaling is independent of α.

Fig. 1 shows simulations results for the TAMSD. If evaluated during the initial time period, $t_a = 0$, the TAMSD for individual trajectories scatter around the ensemble average $\langle \overline{\delta^2} \rangle$, Eq. (12). In contrast, for the aged process ($t_a \gg T$) the ensemble average $\langle \overline{\delta^2} \rangle$ appears much lower than the shown individual trajectories. This is due to the fact that a significant fraction $1 - m_\alpha$ of particles do not move during the entire measurement time. Such trajectories are naturally not visible in a logarithmic plot, while being included in the calculation of $\langle \overline{\delta^2} \rangle$.

In a biological cell, the diffusive motion of a tracer particle is spatially confined, or a charge carrier in a disordered semiconductor experiences a drift force. To address such systems we now determine the TAMSD in the presence of an external potential. We start with a Hookean force $-\lambda x$ and address the more general case below. To that end the continuum approximation of the process is represented by the Langevin equation [34]

$$dx/dn = -\lambda x(n) + \xi(n),$$
(13)

where $\xi(n)$ is white Gaussian noise with $\langle \xi(n_1)\xi(n_2) \rangle = \sigma^2 \delta(n_2 - n_1)$. In other words, $x(n)$ is a stationary Ornstein-Uhlenbeck process. Its increments are Gaussian variables, characterized by the variance $\langle [x(n_2) - x(n_1)]^2 \rangle = \sigma^2[1 - \exp(-\lambda|n_2 - n_1|)]/\lambda$. To calculate the TAMSD, we follow the approach outlined for the free particle, a general approach to compute this and similar

quantities will be provided below. The result reads

$$\left\langle \overline{\delta^2(\Delta; t_a, T)} \right\rangle = \frac{\Lambda_\alpha(t_a/T)}{\Gamma(1+\alpha)} \frac{2K_\alpha \Delta}{T^{1-\alpha}} E_{\alpha,2}\left(-\lambda_\alpha \Delta^\alpha\right) \quad (14)$$

in terms of the generalized Mittag-Leffler function [35], where $\lambda_\alpha = \lambda/\tau^\alpha$. We deduce the limiting behavior

$$\left\langle \overline{\delta^2} \right\rangle \sim \frac{2\Lambda_\alpha(t_a/T)K_\alpha}{\Gamma(1+\alpha)T^{1-\alpha}} \begin{cases} \Delta, & \Delta \ll \lambda_\alpha^{-1/\alpha} \\ \frac{\Delta^{1-\alpha}/\lambda_\alpha}{\Gamma(2-\alpha)}, & \Delta \gg \lambda_\alpha^{-1/\alpha} \end{cases}. \quad (15)$$

Eq. (14) is another special case of Eq. (2). Interestingly the entire dynamics are multiplied by the unique factor Λ_α. Let us now test the generality of this feature.

Consider the time average of some observable $F(x_2, x_1)$ along the trajectory,

$$\left\langle \overline{F(\Delta; t_a, T)} \right\rangle = \int_{t_a}^{t_a+T-\Delta} \frac{\langle F(x(t+\Delta), x(t)) \rangle}{T-\Delta} dt. \quad (16)$$

F may represent moments ($\overline{F} = \overline{\delta^q}, F(x_2, x_1) = |x_2 - x_1|^q$) or the TA of a correlation function. We only require that the jump process $x(n)$ and the function F fulfill

$$\langle F(x(n_2), x(n_1)) \rangle = f(n_2 - n_1). \quad (17)$$

For instance, we have $f(n) = \sigma^2 n$ for the second moment of unbounded motion [cf. (10)], or $f(n) = \sigma^2[1 - \exp(-\lambda n)]/\lambda$ for the TAMSD in an harmonic potential. Condition (17) is fulfilled whenever $x(n)$ is a stationary process (e.g., equilibrated Brownian motion). Alternatively, one may consider a process with stationary increments (e.g., unbounded Brownian motion), when $F(x_2, x_1) = F(x_2 - x_1)$. In these cases we find

$$\left\langle \overline{F(\Delta; t_a, T)} \right\rangle = \int_{t_a}^{t_a+T-\Delta} \int_0^\infty \frac{f(n_a)p(n_a; t, \Delta)}{T-\Delta} dn_a dt,$$
(18)

where $p(n_a; t_a, t)$ is defined in Eq. (3). We obtain

$$\left\langle \overline{F(\Delta; t_a, T)} \right\rangle = C + \frac{\Lambda_\alpha(t_a/T)}{\Gamma(1+\alpha)} \frac{g(\Delta/\tau)}{(T/\tau)^{1-\alpha}}, \quad (19)$$

at short lag times $\Delta \ll T$, with the constant $C = f(0)$. The function g in Laplace space is defined as [25]

$$g(s) = s^{2\alpha-2} \mathscr{L}\{f(n) - f(0); n \to s^\alpha\}. \quad (20)$$

Comparing with our specific results (2), (12), and (15), we identify all relevant terms in TAs: (i) In the limit $\Delta \to 0$, the TA reduces to the constant C, which equals the expectation value of the observable when measured at identical positions. For example, if we study correlations in an equilibrated process, $F(x_2, x_1) = x_2 x_1$, then $C = \langle x^2 \rangle$ is the thermal value of x^2. Conversely, C naturally vanishes if we are interested in TA moments of displacements, $F(x_2, x_1) = |x_2 - x_1|^q$, so it did not appear previously. (ii) The lag time dependence enters
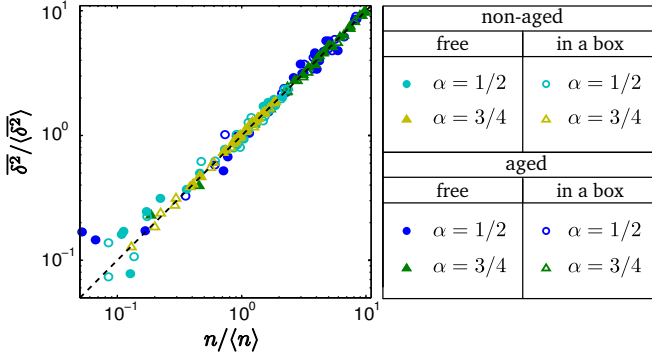
Figure 2: Numerical validation of Eq. (21), for various α, boundary conditions, and t_a (see key). Each point in the graph represents an individual trajectory. Parameters are $\tau = 1$ (a.u.), $\sigma^2 = 1$, $\Delta = 100$, $T = 2 \times 10^6$. t_a is either 0 (non-aged), or for specific α chosen such that $m_\alpha = 0.054$ (aged).

through the function $g(\Delta)$. For example, if $f(n) \sim n^q$, then $C = 0$, and in Laplace space $g(s) \sim s^{\alpha-2-\alpha q}$, which implies $g(\Delta) \sim \Delta^{1-\alpha+\alpha q}$ as in Eq. (12). (iii) The ageing depression function Λ_α only depends on the ratio t_a/T and the parameter α, and due to a factor $T^{\alpha-1}$ any TA converges to the constant C as $T \to \infty$. Note that this dependence on ageing and measurement time t_a and T is universal in the sense that it is indifferent to the specific choice of observable F or model of the jump process $x(n)$, but is directly deduced from the nature of the ageing counting process $n(t)$. Also note that in the Brownian limit $\alpha = 1$, Eq. (19) reduces to $\langle \overline{F(\Delta; t_a, T)} \rangle = f(\Delta/\tau)$, restoring the ergodic equivalence of ensemble and time averages, and the stationarity of the process.

Distribution of TAMSD. Due to the scale-free nature of the distribution $\psi(t)$ of waiting times all TAs of physical observables, e.g. $\overline{\delta^2}$, remain random quantities, however, with a limiting distribution $\phi(\xi)$ for the dimensionless ratio $\xi = \overline{\delta^2}/\langle \overline{\delta^2} \rangle$ [6, 10, 36]. As contributions to time averages of the form (1) occur at time instants when the particle performs a jump, we expect that in the sense of distributions both $\overline{\delta^2}$ and n_a should be equivalent, $\overline{\delta^2} \stackrel{d}{=} cn_a$, for some non-random, positive c. In other words,

$$\xi = \frac{\overline{\delta^2(\Delta; t_a, T)}}{\langle \overline{\delta^2(\Delta; t_a, T)} \rangle} \stackrel{d}{=} \frac{n_a(t_a, T)}{\langle n_a(t_a, T) \rangle}, \qquad (21)$$

for $\Delta \ll T$. We may thus deduce the statistics directly from the underlying counting process. Fig. 2 provides numerical evidence for this argument in the case of a free particle and a particle in a box for several values of α.

The distribution $\phi(\xi)$ for $t_a = 0$ is related to a one-sided Lévy stable law [6]. In the opposite case $t_a \gg T$, combination of Eqs. (21) and (7), yields

$$\phi(\xi) \sim [1 - m_\alpha(T/t_a)]\, \delta(\xi) + m_\alpha(T/t_a)\Gamma(2 - \alpha)$$
$$\times \frac{(T/t_a)^{1-\alpha}}{\Gamma(\alpha)} H_{1,1}^{1,0}\left[\xi \frac{(T/t_a)^{1-\alpha}}{\Gamma(\alpha)} \,\middle|\, \begin{matrix} (2 - 2\alpha, \alpha) \\ (0, 1) \end{matrix} \right], (22)$$

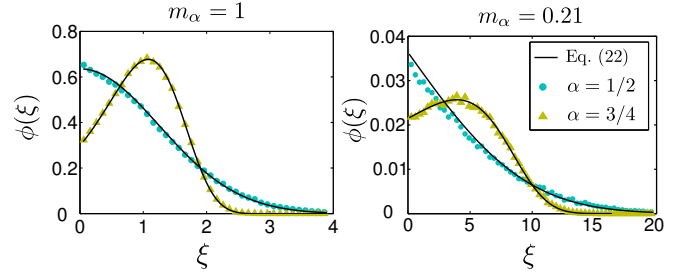Figure 3: Scatter density $\phi(\xi)$ for different α and m_α, see text. Lines: Eq. (7) from [6] (Left) and Eq. (22) (Right). Symbols: Simulations of free CTRW. Note that the area under the curves for the aged process in the right panel is not unity, since the fraction $1 - m_\alpha$ of immobile events is not shown. Same parameters as in Fig. 2.
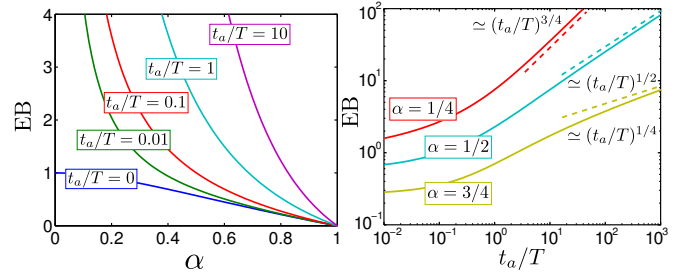

Figure 4: Ergodicity breaking parameter (23) as function of α (Left) and t_a/T (Right). Notice that the non-ergodic fluctuations become larger with increasing t_a.

for $\Delta \ll T \ll t_a$. In $\phi(\xi)$, $m_\alpha(T/t_a)$ is the weight of the continuous part. The probability for not moving during the whole measurement period, ($\xi = 0$), approaches unity as $\simeq (T/t_a)^{1-\alpha}$. If conditioned to measurements with $\xi > 0$, we also find the scaling $\xi \sim (t_a/T)^{1-\alpha}$. In Fig. 3 we demonstrate excellent agreement of Eq. (22) with numerical simulations.

Deviations from ergodic behavior are quantified by the ergodicity breaking parameter [6], for which we obtain

$$\text{EB} = \frac{\left\langle \overline{\delta^2}^2 \right\rangle}{\left\langle \overline{\delta^2} \right\rangle^2} - 1 = 2\alpha \frac{B\left([1 + t_a/T]^{-1}, 1 + \alpha, \alpha\right)}{[1 - (1 + T/t_a)^{-\alpha}]^2} - 1,$$
$$\tag{23}$$

depending only on the ratio t_a/T. At $t_a = 0$ EB reduces to the result of Ref. [6], while for $t_a \gg T$, we find $\text{EB} \sim 2(t_a/T)^{1-\alpha}/[\alpha(1 + \alpha)]$. In the non-aged case EB is bounded, $0 \leq \text{EB} \leq 1$, depending on the value of α only. In contrast we find that EB may *diverge* in the limit $t_a/T \to \infty$. This implies that the non ergodic fluctuations are much larger in the aged regime under investigation. We show the behavior of EB in Fig. 4.

Conclusions. We investigated the effects of ageing on TAs of physical observables. Previous calculations of TAs tacitly neglect the fact that often the preparation of the system and start of the measurement do not coincide.

While this does not cause any problems for ergodic systems with rapid memory loss of the initial conditions, in general this cannot be taken for granted in processes of anomalous diffusion. Here we showed for the case of CTRW with diverging characteristic waiting time that TAs of arbitrary physical observables carry the common factor Λ_α. This ageing depression function is universal in the sense that it only depends on the process age t_a and the measurement time T. All details such as confinement effects enter through a single function, $g(\Delta)$. The structure of this result was shown to hold for a large class of physical observables. We also see that the ageing of the process has a pronounced statistical effect, splitting the population into two: the mobile fraction m_α and the immobile one whose amplitude $1 - m_\alpha$ grows with t_a/T. Knowledge of this effect is significant for the quantitative physical interpretation of experimental data. Finally, since renewal theory is applicable to many systems, our results with minor changes should be relevant more generally, for instance, to the Aaronson-Darling-Kac theorem in infinite ergodic theory or for counting the number of renewals in blinking quantum dots.

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